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DEVELOPMENT OF ION EXCHANGE PROCESSES FOR THE REMOVAL OF RADIONUCLIDES FROM MILK

D. G. Easterly, et al

Environmental Protection Agency
Rockville, Md.

January 1971

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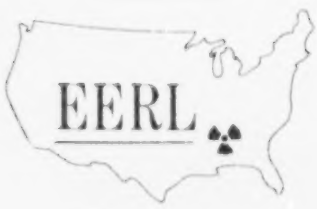

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January 1971

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FOREWORD

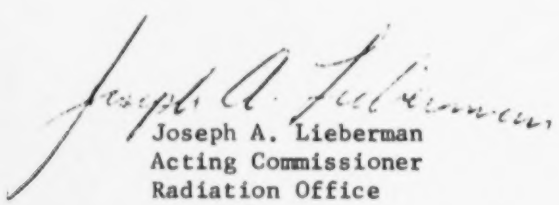
The Environmental Protection Agency has the responsibility of carrying out a national program designed to measure the exposure to the population to ionizing and nonionizing radiation and to assess the radiological quality of the environment.

As one of two regional laboratories within the Environmental Protection Agency, the Eastern Environmental Radiation Laboratory contributes to this program through endeavors in research, applied technology and training. These include development of analytical and radiochemical methodology and development and presentation of courses on the radiological health aspects of ionizing and nonionizing radiation.

The Environmental Protection Agency publishes its findings in Radiological Health Data and Reports (a monthly publication), appropriate scientific journals, and Eastern Environmental Radiation Laboratory (EERL) technical reports.

The technical reports of the EERL allow comprehensive and rapid publishing of the results of intramural and contractor projects. The reports are distributed to State and local radiological health program personnel, Radiation Office and Division technical staff, Radiation Office and Division advisory committee members, university radiation safety officers, libraries and information services, industry, hospitals, laboratories, schools, the press, and other interested individuals. These reports are also included in the collections of the Library of Congress and the National Technical Information Service.

The readers of these reports are encouraged to comment to the Radiation Office on material contained in the reports.

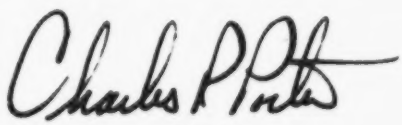


Joseph A. Lieberman
Acting Commissioner
Radiation Office

PREFACE

Since the late fifties work has been conducted to develop methods for removal of radionuclides from milk. In 1960 the Public Health Service, the Agricultural Research Service, and the Atomic Energy Commission became jointly engaged in the development of an ion exchange method for this purpose. These investigations progressed from the laboratory, through the pilot plant, culminating in a full-scale commercial process using fixed-bed ion exchange columns. Laboratory scale and pilot plant investigations were also conducted to evaluate the moving-bed ion exchange system for the removal of radionuclides from milk.

This report reviews some of the research on milk decontamination by ion exchange from the late fifties to June 1969, at which time the milk decontamination project being conducted by the Bureau of Radiological Health of the Public Health Service was discontinued.



Charles R. Porter
Director
Eastern Environmental Radiation
Laboratory

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CONTENTS

	Page
FOREWORD	iii
PREFACE	v
ABSTRACT	ix
1. INTRODUCTION	1
1.1 MILK DECONTAMINATION SYSTEMS	1
1.2 DESCRIPTION OF THE SYSTEMS	2
2. HISTORY	3
3. DESCRIPTION OF PROCESSES	4
3.1 FIXED-BED PILOT PLANT	4
3.2 FIXED-BED COMMERCIAL SCALE PLANT	7
3.3 MOVING-BED PILOT PLANT	8
4. TEST RUNS	12
4.1 LABORATORY SCALE FIXED BED	12
4.2 LARGE SCALE FIXED BED	12
4.3 LARGE SCALE COMBINED ANION-CATION FIXED-BED SYSTEM	14
4.4 PILOT SCALE MOVING BED	14
5. RESULTS	16
5.1 LARGE SCALE FIXED-BED CATION RUNS	16
5.2 LARGE SCALE COMBINED ANION-CATION FIXED-BED SYSTEM	17
5.3 PILOT PLANT MOVING-BED SYSTEM	17
6. ENGINEERING EVALUATION OF THE MOVING-BED UNIT	20
7. RECOMMENDATIONS	23
REFERENCES	27
BIBLIOGRAPHY	29
APPENDIX A -- Summary of Seminar Proceedings	31

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Page

FIGURES

1. Pilot plant equipment used for removing radiostrontium from milk by use of fixed-bed ion exchange columns..... 6

2. Loop of moving-bed pilot plant..... 10

3. Flow diagram of the moving-bed pilot plant equipment used for removing radionuclides from milk..... 11

TABLES

1. Composition of control and treated milk using 1N HCl for acidification..... 18

2. Composition of control and treated milk using 0.75M citric acid for acidification..... 19

ABSTRACT

Work has been conducted on the removal of radionuclides from milk by ion exhange from the late fifties to June 1969. This has progressed from laboratory-scale to a full-scale commercial process where the system was demonstrated to be feasible using fixed-bed ion exchange columns.

For three full-scale combined anion-cation runs, an average of 99.3 percent of iodine-131 and 94.6 percent of strontium-85 was removed. This process resulted in an increase in the total solids, ash, potassium and citrate ions. The taste of the treated milk compared favorably with untreated milk, although some decrease in flavor score was noted.

Good radionuclide removal was obtained using the pilot plant moving-bed system. These studies, although not extensive, showed 89, 99, and 99.5 percent removal for strontium, iodine, and cesium, respectively. The limited investigations of cesium removal showed this system to have much greater potential than the fixed-bed system. For complete evaluation of this system, additional research would be needed.

The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as either an actual or implied endorsement of such products by the Department of Health, Education, and Welfare, Public Health Service.

THE AUTHORS

During the time of the work described in this report, Mr. D. G. Easterly, Mrs. I. B. Brooks, and Mr. J. K. Hasuike were chief, biologist, and engineer, respectively, of the Countermeasures Unit, Southeastern Radiological Health Laboratory, Montgomery, Ala.; and Mr. C. L. Weaver was Chief, Division of Environmental Radiation, Bureau of Radiological Health, Rockville, Md. They are all now assigned to the Radiation Office, Environmental Protection Agency.

DEVELOPMENT OF ION EXCHANGE PROCESSES FOR THE REMOVAL OF RADIONUCLIDES FROM MILK

1. INTRODUCTION

In the past, nuclear bomb testing by the nuclear powers released strontium-90, cesium-137, and iodine-131 into the atmosphere. Deposition of radionuclides on the land led to a transfer chain to the milk food supply. With the cessation of atmospheric testing in 1962 by those countries signatory to the test ban treaty, the situation has improved. Other countries, however, have continued to test nuclear devices in the atmosphere. The resumption of large-scale atmospheric nuclear testing or a nuclear accident near a milk producing area might cause unacceptably high radionuclide levels in fresh milk. If radioactive contamination occurred in a limited area for a relatively short time, several alternatives for protective action exist. These include, among others, providing stored feed for cattle until decay of the nuclides has reduced the radioactivity available for incorporation in milk to an acceptable level, supplying the area with fresh milk from another source, or diverting the milk to other uses.

1.1 MILK DECONTAMINATION SYSTEMS

In the event of radiological contamination of milk over a wide geographical area, a practical system for removing long half-life radioactivity from large quantities of fluid milk would be an important means of continuing the source of fresh milk for the consumer market. Under this condition the public should have readily available a means of reducing internal radiation exposure from radionuclides in milk.

The Public Health Service, Bureau of Radiological Health, recognized this need and began to investigate methods which would effectively remove the biologically significant radionuclides from milk. The Bureau of Radiological Health has been involved in several approaches toward milk decontamination. The purpose of each of these activities was to investigate methodology and to test this methodology so that the industry would have the required technology and be prepared to decontaminate milk if there is a problem in the future.

Although several means of milk decontamination have been explored, the use of ion exchange resins has received by far the greatest attention. There are two basis types of equipment used in ion exchange work--those utilizing the fixed bed and those using the moving bed systems. The fixed-bed system has been developed through the commercial stage, while the moving bed system has had limited pilot plant investigations.

Even though the fixed-bed system had been thoroughly studied, the moving bed equipment manufacturers believed their system offered substantial benefits. To explain the moving bed system and its advantages, Mr. Jack D. Ferner, President, Chemical Separations Corporation, Oak Ridge, Tenn., presented testimony at a hearing August 27, 1963, before the Subcommittee on Research, Development, and Radiation of the Joint Committee on Atomic Energy. Mr. Ferner testified that the methodology for removing radioactive fallout materials from milk had been developed by his company. He stated that the fixed-bed ion exchange system "makes two chemical additions to the milk which changes the composition and balance of the major salts." According to Mr. Ferner it also "causes a noticeable taste difference, and perhaps worst of all, it adds another radionuclide, potassium-40."

Mr. Ferner also stated that his system was effective in removing the contaminants, reasonable in cost, practical to use on a large scale in commercial dairy operations and caused no deleterious side effects to the milk.

1.2 DESCRIPTION OF THE SYSTEMS

In the continuous or moving-bed system which Mr. Ferner discussed, the resin bed is pulsed or circulated counter-current to the milk flow in a loop divided into specific compartments. The milk treatment and regeneration of the resin occur simultaneously in separate compartments, and the milk flow is interrupted only intermittently to allow fresh resin to be pulsed into the milk treatment section. When the system pulses, a portion of the exhausted resin in the milk treatment section is moved into the regeneration section of the loop, regenerated, rinsed, and then recycled to the working portion of the loop where it is again ready to repeat the entire ion exchange operation.

In the fixed-bed ion exchange milk decontamination system the milk is fed through a resin bed until the resin is exhausted and then the milk is diverted to another resin bed while the previous exhausted system is being regenerated and sanitized.

Chemically the two systems are identical, but they differ in mechanics. Compared to a fixed-bed system, the moving-bed system (according to its manufacturer) has three major advantages: (a) it

is continuous, (b) it requires smaller volumes of resin, and (c) it incurs less milk loss. Other advantages include the use of less regenerant per pound of milk processed and higher sustained removal of radionuclides through the continuous availability of freshly regenerated resin in the milk treatment section of the loop. The latter factor may be especially significant in the case of cesium, for which a decrease in removal efficiency is experienced after approximately 12 bed volumes (bv) of milk have been treated in a fixed-bed system.

2. HISTORY

In the late fifties Easterly et al.[1,2] showed that the radioactivity in milk could be reduced by an ion exchange treatment. In 1959 Crosslett and Watts[3] reported 98 percent removal of in vitro labeled radiostrontium from milk that was passed down a column of zeocarb-225 resin in the calcium form. The above procedure reduced the levels of radioactivity in milk, but also altered the ionic composition of the treated milk. In an attempt to avoid disruption of the ionic balance of the milk, Migicovsky[4] used an exchange resin in a mixed ionic form. When milk labeled in vitro with radiostrontium was treated in batches with this charged resin, 79 percent of the radionuclides were removed by the first treatment but rose to 97 percent by the fourth treatment.

At the USDA Dairy Products Research Laboratory, Beltsville, Md., a fixed resin bed in a column was used and the mixture of ions on the resin was modified. About 50 percent of the strontium in milk was exchangeable at the normal milk pH and could be increased to 90 percent or more by reducing the pH to 5.3 or less.

In 1960, a pilot plant was constructed at Beltsville, Md., under the sponsorship of the Public Health Service, the Department of Agriculture, and the Atomic Energy Commission for further refinement of the process with a view to adapting it to commercial use. Also at Beltsville in 1961, the Department of Agriculture and the Public Health Service began conducting laboratory investigations of a continuous ion exchange system for milk decontamination.

To evaluate the fixed-bed process on a commercial scale, the Government contracted in 1963 with a commercial milk processor (Producers' Creamery Company, Springfield, Missouri) for the design,

construction and operation of a full-scale processing plant based on the pilot plant design.

While the strontium-90 removal system was being evaluated in the pilot and commercial-scale plants, laboratory investigations were being conducted for the development of methods to remove radioactive iodine from milk by ion exchange. By the time the full-scale tests on the strontium-90 removal plant were completed, full-scale operation for the removal of radioanions by ion exchange also appeared practicable. Therefore, under sponsorship of the Public Health Service and the Department of Agriculture, in 1965 a contract was made with the same commercial milk processor to design and build a full-scale system to remove iodine-131 from milk and to incorporate the system into the existing full-scale plant. The contract also included testing the combined plant to confirm the practicability of the strontium-iodine removal process. Commercial feasibility was demonstrated and a food additive petition for emergency use of this process has been submitted to the Food and Drug Administration for approval.

A memorandum of understanding was executed between the Department of Health, Education, and Welfare, Public Health Service, and the Department of Agriculture, Agricultural Research Service, on May 13, 1964, for the development of a system for removal of radioactive contamination from fluid milk by the use of a moving resin bed. In April 1964, based on Mr. Ferner's testimony, the Joint Advisory Committee on Removal of Radioactivity from Milk endorsed the proposal to proceed with a contract for the continuous contactor pilot plant.

3. DESCRIPTION OF PROCESSES

3.1 FIXED-BED PILOT PLANT

Figure 1 shows the pilot plant equipment used for removing radiostrontium from milk by use of fixed-bed ion exchange columns. This flow sheet is sufficient to follow the characteristic steps in the process; however, some of the important equipment units are not indicated, particularly the control panel, milk filters and "in-line" pH electrodes. Four 200-gallon regenerant tanks provided for the reuse of three-fourths of the salt solution (only three tanks are shown in figure 1). The pilot plant was designed for a capacity of 100 gallons per hour. The process included the following operations:

Initial Resin Preparation. The resin is regenerated initially with a mixed salt solution of calcium, magnesium, potassium, and sodium chlorides. The composition of the solution used was (in grams per liter): $CaCl_2 \cdot 2H_2O$, 535; KCl, 23.1; NaCl, 8.5; and $MgCl_2 \cdot 6H_2O$, 15.1. These quantities were selected so the resin would be equilibrated with respect to the major milk cations, thus assuring a minimum of

change in the mineral composition of the resin-treated milk. About 10 resin-bed volumes (rbv) of the fresh salt solution are usually sufficient for the initial regeneration; the exact quantity depends on the temperature and flowrate of the regenerant.

Rinsing of Excess Salt Solution. All excess salt solution should be thoroughly rinsed from the column with water.

Acidification of Milk. The cold, raw, whole milk is adjusted to 10°C., then acidified to pH 5.3 by an acid-metering pump with a solution of 0.75M citric acid (fig. 1). The milk is then filtered to remove any fine material which might cause plugging in the resin column.

Flow of Milk Through the Resin Column. Referring to figure 1, the milk would flow through Column I by opening valves 2 and 6. After a period of time (up to 8 hours of continuous flow) the milk may be diverted to Column II by closing valves 2 and 6 and opening valves 3 and 7. Column I may then be rinsed, sanitized, and regenerated. For practical use either of two flowrates is suggested: 1/8 or 1/16 rbv per minute. For a capacity of 100 gallons per hour these rates require backwashed and settled resin volumes in the column of 13.35 or 26.7 gallons, respectively. The resin inventory per 8 hours of processing time and regenerant requirements per unit volume of milk will be the same in either case. Thirty rbv of milk flowing through a column require 8 hours at 1/16 rbv per minute or 4 hours at 1/8 rbv per minute.

Neutralization and Processing of Treated Milk. The effluent milk from the columns is neutralized by continuous in-line addition of a solution of 1.5 N KOH or with other bases. It may then be pasteurized and homogenized in the usual manner.

Cleaning and Sanitizing the Resin. After milk is diverted to Column II (fig. 1), Column I may be rinsed by opening valves 11, 9, 1, 5, 13, and 12. To wash upflow through the column with a detergent solution valves 11 and 9 are closed; valves 14, 10, 5, 1 and 18 are opened.

Regeneration of the Resin. This procedure is carried out with the same salt solution used for the initial preparation. The chief purpose of regeneration is to remove the radiostrontium from the resin, but it also maintains equilibrium with respect to the minerals in milk. It is economically desirable to reuse as much of the regenerant as possible. The process provides for reusing three-fourths of this solution. The first fraction (one-fourth of the total) through the column is discarded. Each of the three remaining frantions is pumped

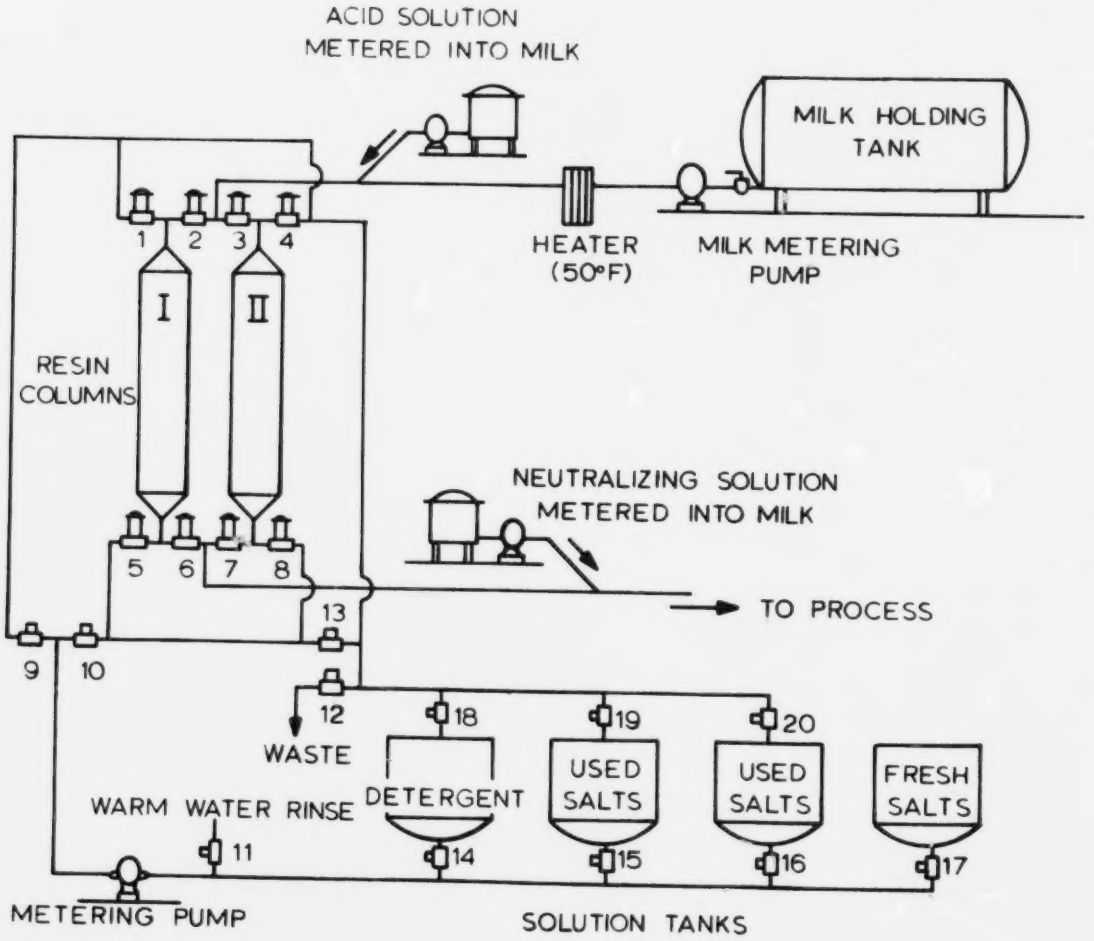

Figure 1. Pilot plant equipment used for removing radiostrontium from milk by use of fixed-bed ion exchange colums.

into separate tanks, which are used for subsequent regeneration in the sequence in which they are recovered. The fourth tank in the series is filled with fresh solution equal in volume to the amount discarded from the first tank in the series.

The regenerant is passed downflow through the column by opening the appropriate valves indicated in figure 1. For this process about 16 rbv of regenerant are required, 4 rbv of which are fresh solution. Thus, 1 volume of salt solution is required for each 7.5 volumes of milk. The effectiveness of radionuclide removal from resin is increased by heating the regenerant solution. A further advantage of heating this solution is in the reduction of bacterial growth in the resin columns. The advantages gained may be partially offset by the expense of heating.

3.2 FIXED-BED COMMERCIAL SCALE PLANT

The Government contracted in 1963 with a commercial milk processor (Producers' Creamery Company, Springfield, Missouri) for the design, construction, operation, and evaluation of a full-scale processing plant based on the pilot-plant design. The pilot-plant system could be considered a prototype of the commercial scale process.

The commercial plant was designed using 3A standards and following the U.S. Public Health Standard Ordinance and Code wherever applicable. The plant had a rated capacity of 5,700 liters per hour of milk for an 8-hour run. Two 22,000-liter storage tanks were utilized, one for raw milk prior to processing, and one to receive pasteurized milk after removal of strontium-90.

The raw milk was pumped with a positive pump from the storage tank at the rated capacity, through a plate heat exchanger to bring it to about 13° C. To achieve rapid and complete mixing of milk and acid solution, the citric acid solution (20 percent) was pumped with a variable-drive, positive pump (57 liters/hr) through plastic tubing into the eye of a centrifugal line mixer. The volume of acid injected was regulated by a pH controller to maintain the pH of the milk at 5.35. The acidified milk then passed through one of two inline filters to one of two stainless steel columns. The filters were alternated if required, because of increasing pressure, by means of pressure-activated valves. These resin columns were custom-made, 107 cm diameter by 244 cm high (488 cm with legs) and contained 1,325 liters of IR-120 resin.

After passing downflow through the resin bed, the milk was neutralized to pH 6.8, using 6 percent potassium hydroxide solution fed

by a variable-drive positive pump (210 liters/hr) into the eye of a centrifugal pump. After pH adjustment, the milk was allowed to accumulate in a surge tank for about 15 minutes prior to pumping with a positive pump through a plate heat exchanger to pasteurize the milk. It was then cooled and stored cool until it was spray-dried. Although the processed milk was satisfactory organoleptically, it was all diverted to animal food use.

The capacities, in liters, of the polyethylene-lined, steel milk tanks were 9,500 for the regenerant, 800 for the citric acid, and 2,000 for the potassium hydroxide solution. The entire system was fully automated except for regeneration sequencing, which was manually operated to reduce the number of regenerant tanks required. The processing equipment was installed in an area 7.3 by 18.3 m, in a room with a ceiling height of 4 m.

Additional columns were added for the combined process of removing both strontium-90 and iodine-131 from fluid milk. These anion columns were constructed of mild steel and completely lined with a continuous layer of polyethylene 6.3 mm thick. The column was 224 cm high with an inside diameter of 65 cm. Sight glasses were placed 118 cm and 195 cm from the bottom of the column. Pyrex glass balls 2.5 cm in diameter were placed in the bottom of the column with 1 cm diameter glass balls layered on top of the 2.5 cm size to form a support for a 60-mesh, monofilament nylon screen. Dowex 2 x 8 20-50 mesh was placed on top of the nylon screen. The volume of resin was 265 liters (one bv). After modifications for the combined process the complete plant was operated manually since automation of the changes would have added substantially to the cost of the project.

3.3 MOVING-BED PILOT PLANT

The moving-bed pilot plant unit, which was designed to have a capacity of 100 gallons of milk per hour, consists of two fixed-bed columns for radioactive anion removal and two moving-bed loops comprising the continuous ion exchange contactors. One loop was to be used to remove radioactive cations and the other, the anion loop, was for milk neutralization.

Each of the two moving bed loops is composed of four main compartments. Thus, in the first compartment of the cation loop radionuclides are removed from the milk (in the anion loop this section is used for milk neutralization); in the second compartment the resin is rinsed free of milk; in the third section the resin is regenerated; and in the final compartment the resin is rinsed free of the regenerant solution.

The unit is self-contained (except for power supply) and is designed for mounting on a standard 40' x 8' truck trailer to make it readily transportable.

The simplified flow diagram of the pilot plant is shown in figure 2. From storage, the milk flows by gravity to milk pump 1 where it is pumped through the anion fixed-bed column, acidification injection point, line mixer, and into the open surge tank. The milk then flows by gravity to milk pump 2 where it is pumped sequentially through an inline filter, cation moving bed working section, anion moving bed working section, and finally into storage.

The design called for alternating the use of two anion fixed beds to remove iodine-131, an acidification system for lowering the milk pH, a stainless steel loop for the cation moving resin bed to remove strontium-90 and cesium-137, and a glass loop containing the anion moving resin for milk neutralization.

Regeneration of the resin for the moving-bed studies was carried out in the same manner as for the fixed-bed system. The cation resin used, however, was Dowex 50WX-8 instead of IR 120.

During operation the system is either in pulse mode (resin movement) or a run mode (milk and resin treatment). The pulse mode can be timed from 0 to 30 seconds and also can be left on pulse indefinitely. The run mode can be timed from 0 to 60 minutes.

When the system cycles to the pulse mode, all operations pertaining to the run mode shut down; (see figure 3) resin valve A (RVA) closes; RVB, RVL, RVC, and RVD are opened; the pulse pump pulses the resin. Before the end of the pulse mode, the pulse bleeder valve opens to drain the system of excess water. The opening of the bleeder valve can be delayed anywhere from 0 to 6 seconds. At the end of the pulse mode the system shifts back to the run mode. Resin valves RVB, RVL, RVC and RVD close while RVA opens to allow the pulsed resin to fall back into the pulse chamber. After a delay of a few seconds, all valves labeled "Z" open.

Z_a = when the solubridge senses milk at probe a, the drain valve will close and the MILK OUT valve will open.

Z_b = after each pulse the conductivity is so high at probe b that the solubridge will close the rinse-out valve (Z_b) to force the salt water interface back to probe b, after which the rinse-out valve will open to allow further rinsing.

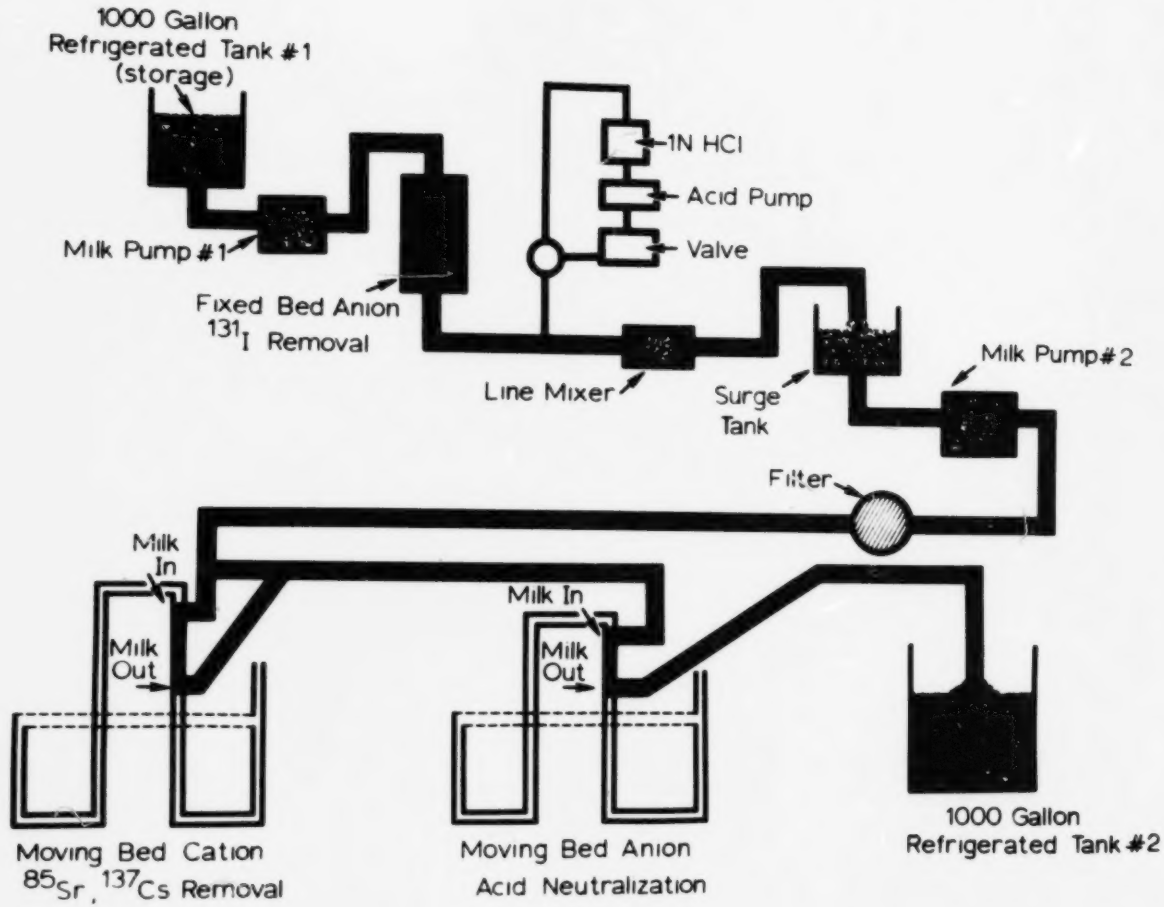

Figure 2. Flow diagram of the moving-bed pilot plant equipment used for removing radionuclides from milk.

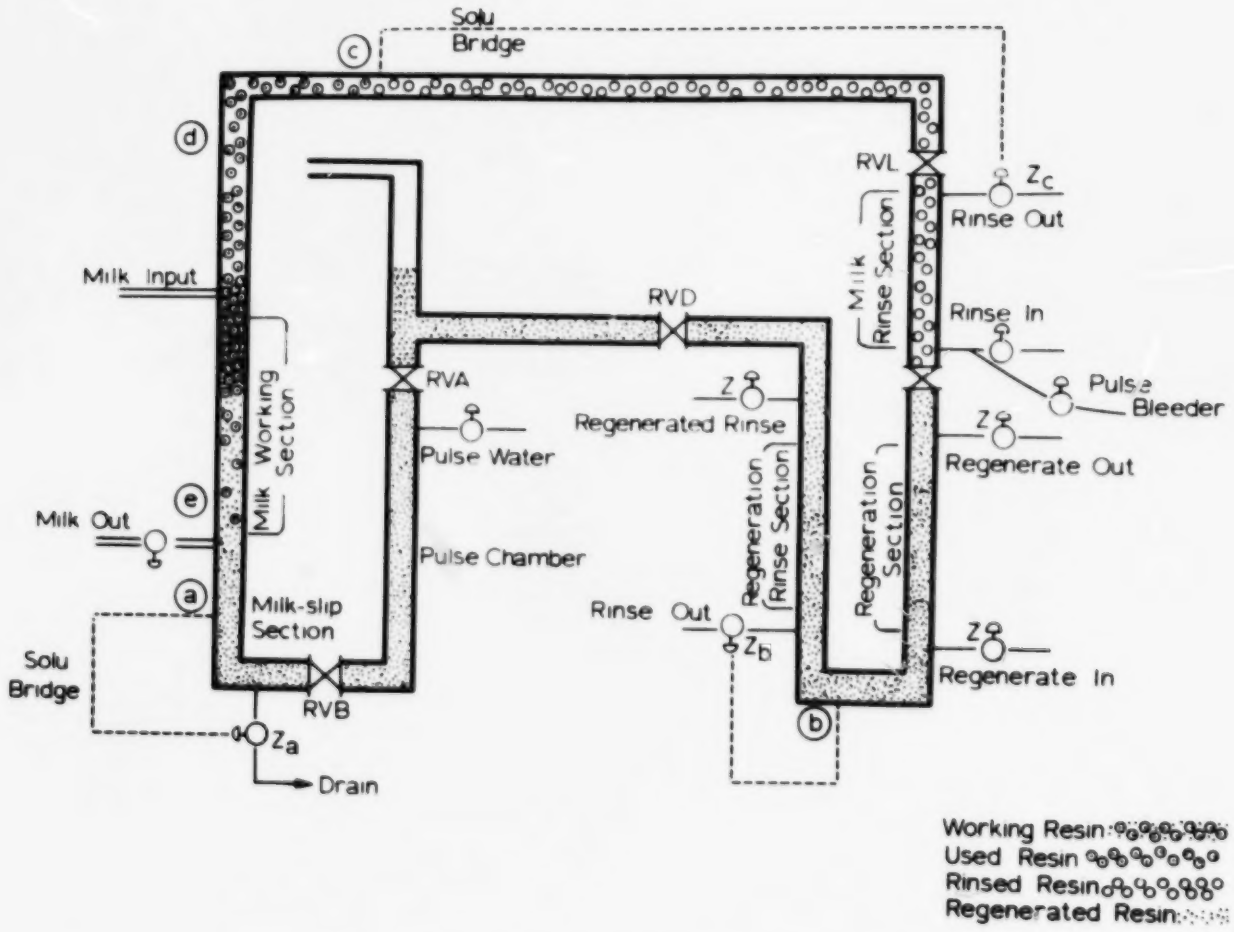

Figure 3. Loop of moving-bed pilot plant.

Z_c = after each pulse the solubridge probe c will be immersed in milky resin. RVL remains open while the rinse-out valve (Z_c) closes to force the milk-water interface back to probe c.

The milk-resin contact area is confined to the milk working section which is controlled by and located between solubridge probes a and c.

During pulsing, an increment of the milk-contacted resin is pulsed toward the milk rinse section where hot water (65.5°C) rinses the resin free of milk residues. Subsequently, the rinsed increment of resin is pulsed into the regeneration section for recharging and from there to the regenerant rinse section. Eventually this fully recharged increment of resin will return to the milk working section to complete the cycle.

4. TEST RUNS

4.1 LABORATORY SCALE FIXED BED

Practically all of the research carried out during the development of the fixed-bed cation process at Beltsville, Maryland, was done by using strontium-85 because the method for its assay is simple and rapid compared to that for strontium-90. Levels of about 1 μCi per quart of milk were normally used for removal studies. These levels of strontium-85 were about 40,000 times the environmental levels of strontium-90. The average removal of strontium for 30 resin bed volumes of milk was approximately 95%. Only small changes in the flavor of the resin-treated milk were observed. The strontium-85 was determined by gamma spectroscopy using a 3-inch NaI (Tl) well crystal, single-channel analyzer system.

4.2 LARGE SCALE FIXED BED

Fresh Grade A whole milk standardized to 3.6% butterfat or skim milk was delivered to the plant for each run. The system was started on 2°C water, the resin bed cooled to 13°C, and the preheater regulated to maintain the milk temperature at 13°C. Milk was then circulated through the pH electrode system and the preheater, back into the raw milk storage tank until both the temperature and the pH controlling system became stabilized. The pH of the milk was verified by laboratory measurements. Milk was then diverted to forward flow through the column, the neutralizing system, the surge tank, the pasteurizing system, and finally into a storage tank.

At the completion of each milk run, water was introduced into the system to rinse the resin. After a preliminary rinse, the flow was reversed and a nonionic detergent added to aid in removing milk solids from the resin. Temperature of the water was increased until the column effluent reached 48°C. After the detergent wash was completed the column was rinsed at 48°C and finally brought to a temperature above 82°C. Not more than 24 hours before the next run, the column was rinsed with water before regeneration was initiated.

The last portion (4.5 or 9 bed volumes, depending on flow rate and milk volume) of regenerant solution used on the previous run was passed through the column at 60°C at a flow rate of about 75 liters/min. This portion of the regenerant was directed to waste. The freshly prepared portion (4.5 or 9 bed volumes) was heated to 60°C, passed through the column, and stored for the succeeding run. The column was then rinsed with water until all traces of regenerant salts were removed and chilled to 13°C with 2°C water immediately before using.

For each run, the raw milk was shipped in tankers. Samples were taken from each arriving tanker, from pipe lines after leaving the columns, and from the pasteurized processed milk in the tankers as they left the plant.

Radiological assays of the milk were made by Controls for Radiation, Inc., Cambridge, Massachusetts, following the procedure of Martell[5] for the rapid assay and that of Harley[6] for the longer assay.

Analysis of the milk protein was determined by a semimicro-Kjeldahl[7] method and lactose by means of a polarimeter.[8] Citric acid,[9] phosphorus,[10] and iron[11] were determined colorimetrically. Sodium and potassium were determined by flame photometry.[12] Total calcium and magnesium were determined by direct titration with EDTA.[13] Calcium was then determined by permanganate titration of calcium oxalate,[11] and magnesium estimated by difference. Chlorides[14] and titratable acidity[15] were determined by titration. Freezing points were obtained with the Fiske cryoscope.[11] Curd tensions were made using the Cherry-Burrell curd tension meter following ASDA procedures.[16] Fat and total solids were determined by the Mojonnier Tester.[17] Ash was determined at 550°C.[11] A Beckman Model G and a Beckman Zeromatic were used for estimating pH.

Bacteriological analyses were made according to procedures described in Standard Methods.[15] Standard plate counts were made at 32°C. Violet red bile agar and medium 110 were used to enumerate coliform and staphylococci, respectively.

All determinations were made in duplicate or quadruplicate.

4.3 LARGE SCALE COMBINED ANION-CATION FIXED-BED SYSTEM

Prior to the first combined anion-cation run, the Dowex 2 x 8 (anion resin) was conditioned with NaOH and HCl before charging with the anion regenerant solution. The composition of the solution used was (in mM per liter): Na_2HPO_4, 100 $PO_4^{\equiv}$; $Na_3C_6H_5O_7 \cdot 2H_2O$, 185 $C_6H_5O_7^{=}$; NaCl, 125 Cl^-. As in the case of the cation regenerant, these quantities were selected so that the resin would be equilibrated with respect to the major milk anions, thus assuring a minimum of change in the composition of the resin-treated milk.

After processing the milk, the anion exchange column was rinsed by backwashing with 50°C. water until all visible traces of milk disappeared. The column was then backwashed with 5 bv of a non-ionic detergent (0.1 percent Triton X-100) and rinsed with 25 bv of water to remove the detergent. Stripping was conducted with 48 bv of 2 N HCl at a flowrate of 0.2 bv per minute. Regeneration was accomplished by treating the resin with 23 bv of the anion regenerant solution. The anion resin was stored in the column with an HCl solution (pH 1.0) to control microbial growth until the next run.

Analytical procedures for physical, compositorial, bacteriological, and organoleptic changes were essentially the same as those for the cation runs. Radiological assays for iodine-131 and strontium-85 were determined by gamma spectroscopy.

The cation column procedure was the same as for previous cation runs described in section 3.2.

4.4 PILOT SCALE MOVING BED

The moving bed ion exchange unit was delivered to the Southeastern Radiological Health Laboratory on April 12, 1967, and operational testing of the unit was initiated in August 1967. Twenty-five runs were made with the unit: 10 tap water, nine unspiked milk, and six in vitro spiked milk samples. After the tap water and unspiked milk runs, improvements were made on the acidification system which had been erratic during preliminary testing at the Chemical Separations Corporation. Modifications of the system involved the installation of a three-way acid valve and rewiring of the acid pump for continuous operation. Defective pH probes were replaced and internal adjustments were made to align the automatic pH meters. Tap water acidification runs were made with citric acid until effective working conditions were established. Small quantities of milk were then tested for satisfactory results.

Following the preliminary modifications and testing, the moving-bed unit was more completely tested by passing milk through the entire system. The nine unspiked milk runs through the system (as observed through the partial glass anion loop) revealed the following:

— entrapped air in the milk work section,

— introduction of air into the milk rinse section,

— insensitivity of milk-water interface probes,

— regenerant channeled along the sides of the column, and

— resin displacement (disruption of the resin bed).

Similar behavior was assumed to have occurred in the cation loop in which direct observations could not be made because of its stainless steel construction. The air entrapment was eliminated in the anion loop by inserting a tee with a manual bleed-off valve in the top section of the loop. The Beckman solubridge controllers apparently lacked the capability of controlling the milk-water interfaces. Milk fat probably built up around the probes and caused the bridge to malfunction, since sensitivity adequate at the start of a run declined with time. The regenerant channeling could be controlled by carefully regulating the rinse and regenerant flow rates. Resin displacement, however, is inherent with the folded loop design and must be tolerated with this system. The loop neutralization process was not successful and needs further research for improvement.

Although problems such as loop neutralization and resin displacement still remained to be solved, runs with in vitro spiked milk were conducted and removal efficiencies were determined for iodine-131, strontium-85, and cesium-134. The iodine-131 removal studies were done with an anion fixed bed; the strontium-85 and cesium-134 studies were made with a cation moving-bed loop.

The partial glass loop originally designed for neutralization was converted to all glass, filled with cation resin, and used in these studies to observe the resin movement and related characteristics. The glass loop, however, which has only one-third the capacity of the stainless steel loop necessitated reducing the flow rate from 100 gallons per hour (gph) to approximately 35 gph.

For each of six spiked runs about 200 gallons of grade A raw milk were purchased from a local dairy. The milk was received in a 1000-gallon, refrigerated storage tank. All six runs were spiked in vitro with approximately 2000 pCi/l each of iodine-131 and strontium-85. Runs 5 and 6 were also spiked with cesium-134. After spiking the milk with the radionuclides an equilibration time of 24 hours was allowed before each run.

From storage the milk was pumped to the fixed bed anion column and passed downflow at a rate of 100 gph. After leaving the anion column the milk was pumped through an inline acid mixer where the pH

of the milk was lowered to about 5.2 with 1 N HCl in the first two runs and with 0.75 M citric acid in the other four runs. The acidified milk was pumped into a surge tank and held until it could be pumped through an inline filter for removal of localized precipitation before entering the milk working section of the cation loop. The milk flow rate through the loop was 35 gph. From the cation loop, the milk went to storage (1000-gallon refrigerated tank).

The moving bed was pulsed for 12 seconds at 10-minute intervals during each of the runs which were from 3 to 6 hours' duration. Samples of milk were taken after (a) the anion treatment and prior to the cation treatment, (b) from the cation loop both before and after pulsing, and (c) from the final product.

A three-channel gamma spectrometer was used to determine the radionuclide concentration in the samples before and after treatment by measuring the counting rates of iodine-131 at 0.364 MeV, strontium-85 at 0.51 MeV and cesium-134 at 0.79 MeV. A 3 x 3 matrix was determined and applied to correct the counting rate of each radionuclide.

The samples were also assayed for the major cations (Ca^{++}, K^{+}, Na^{+}, Mg^{++}) and anions (Cit:P:Cl). Total solids were determined to measure the dilution that occurred in the processed milk. Standard plate and psycrophile counts were made on selected samples. Control and processed samples (which had been neutralized with 1 N KOH) were pasteurized for taste evaluation from one run. Bacteriological and total solid studies were conducted as described in the 12th edition of Standard Methods for the Examination of Dairy Products.[15]

The major cations were determined by atomic absorption spectroscopy. Citrates and phosphates were determined colorimetrically. Chloride content was determined by titration.

5. RESULTS

5.1 LARGE SCALE FIXED-BED CATION RUNS

Efficiencies of strontium-90 removal from milk from seven 45,400-liter runs and one 90,000-liter run varied from 86.0 to 96.9% and averaged 91.7% for a total of 413,000 liters of milk processed.

There was an apparent decrease in protein, fat, lactose, and certain other constituents in the processed milk. Sodium, potassium and citrates increased, whereas calcium and magnesium decreased with increasing bed volumes of milk processed.

The principal difference between the raw and the processed milk from a compositional standpoint was due to the addition of approximately 0.4 percent of potassium citrate. In the six whole milk runs there was an average increase in citrate ion of 2,357 $\pm$ 584 parts per million, or approximately 0.12 percent, resulting in a total increase of 0.36 $\pm$ .08. This figure compares well with the 0.36 $\pm$ 0.8 percent increase in total solids. The potassium citrate content caused a reduction in curd tension.

A significant increase in titratable acidity and specific gravity and a significant decrease in freezing point were observed. Results of bacterial counts indicated that the columns could be operated under satisfactory sanitary conditions.

5.2 LARGE SCALE COMBINED ANION-CATION FIXED-BED SYSTEM

For three full-scale combined anion-cation process runs, an average of 99.3 percent of the iodine-131 and 94.6 percent of the strontium-85 was removed. This process resulted in an increase in the total solids, ash, potassium, and citrate ions; however, slight increases and decreases were observed for some components. The composition of the processed milk was approximately the same as that obtained from the cation column only. There was a decrease in flavor score from 37.2 to 35.7 because of processing. Fluid, untreated milk dropped in score from 37.4 to 35.4 after drying. Ion exchange treated milk dropped from 35.9 to 34.0 after drying.

5.3 PILOT PLANT MOVING-BED SYSTEM

The percent removal of radionuclides is shown in tables 1 and 2. The average percent removal for these isotopes is considered good and in agreement with previous works. Greater removal of strontium was obtained with the citric acid acidification than with HCl. This agrees with previous results reported by Easterly et al.[18] The limited investigations of cesium removal showed this system to have much greater potential than the fixed-bed system.

Data from chemical analyses of the control and treated milk samples are shown for the HCl (table 1) and citric acid (table 2) acidification runs. The high chloride content in runs 1 and 2 is attributed to the addition of HCl, and the high citrate content in runs 3, 4, 5, and 6 is attributed to the addition of citric acid in the milk acidification process. The K^+ content would have been higher had the milk been neutralized with KOH.

Results of the very limited bacteriological studies are shown in tables 1 and 2. The runs made were neither conducive to nor designed for typical bacterial studies, but milk samples were plated to give some indications of what to expect of the moving-bed system operated as

Table 1. Composition of Control and Treated Milk Using 1 N HCl for Acidification

Analysis	Run No. 1		Run No. 2		Average 1 and 2		Difference
	Control	Treated	Control	Treated	Control	Treated	
Total solids, percent	12.65	12.13	12.63	11.82	12.39	11.98	-0.41
Citrate ppm	1,550	1,616	1,570	1,603	1,560	1,610	+50
Cl ppm	1,046	1,720	1,064	1,781	1,055	1,751	+696
Inorganic P. ppm	730	643	706	710	718	677	-41
Ca	1,096	1,149	1,112	1,378	1,104	1,264	+160
Mg ppm	128	118	130	150	129	134	+5
K ppm	1,552	1,440	1,480	1,068	1,516	1,236	-280
Na ppm	584	542	540	374	562	458	-104
pH	6.46	5.34	6.54	5.28	6.50	5.31	
MPN	180,000	160,000	87,000	39,000			
PBC	38,000	23,000	5,400	1,600			
Percent strontium removal		72.1		80.5		76.3	
Percent iodine removal		99.0		99.2		99.1	
Run time	3 hours		3-1/2 hours				
Charging preliminary	FB charged Cation charged		no				

FB - Fixed Bed.

Table 2. Composition of Control and Treated Milk Using 0.75 M Citric Acid for Acidification

Analysis	Run No. 3		Run No. 4		Run No. 5		Run No. 6		Average Runs Nos. 3-6		Difference
	Control	Treated	Control	Treated	Control	Treated	Control	Treated	Control	Treated	
Total Solids percent	12.60	11.61	12.84	12.31	12.27	11.20	12.07	11.80	12.45	11.73	-0.72
Citrate ppm	1,990	4,992	2,000	4,360	1,600	3,287	1,750	3,478	1,835	4,029	+21.94
Chloride ppm	1,039	763	975	1,072	975	641	1,010	979	999.7	864	-135.7
Inorganic P ppm	717	688	648	635	687	650	622	599	669	643	-26
Ca ppm	1,148	1,309	1,246	951	1,310	1,479	1,300	1,432	1,251	1,293	+42
Mg ppm	130	139	122	194	99	122	100	94	113	137	+24
Na ppm	546	493	526	536	500	456	468	440	510	481	-29
K ppm	1,496	1,120	1,600	1,721	1,540	1,293	1,590	1,248	1,556	1,346	-210
pH	6.56	5.26	6.56	5.35	6.50	5.20	6.50	5.25	6.53	5.27	-
MPN	73,000	167,000	-	-	70,000	139,000	64,000	78,500	-	-	-
PBC	5,000	9,000	-	-	16,000	34,000	23,000	20,000	-	-	-
Removal:											
Percent Iodine		98.6		99.3		99.3		99.1		99.1	
Percent Strontium		96.5		97.2		92.6		95.2		95.4	
Percent Cesium		-		-		99.3		83.7		91.5	
Run Time	6 hrs		6 hrs		5-1/2 hr		5-1/2 hr				
Charging	Cation (FB) Anion no		Anion precharged Cation charged MB		FB no cation from previous run		Anion regenerated Cation as is				

FB - Fixed Bed.
MB - Moving Bed.

described. No definitive conclusions can be drawn until bacterial studies are made under typical operating conditions, but indications are that microbial control will not be difficult.

The total solids results show that a 3 percent dilution occurs when 0.75 M citric acid is used for milk acidification. These values would also be higher had the KOH neutralization procedure been incorporated.

The samples that had been neutralized and pasteurized for taste evaluation were tested by four people. Three of the four did not distinguish the control from the other two samples. The fourth person, however, detected a slight difference between the control and the two treated samples but did not find the treated samples objectionable.

Other research on a laboratory scale model of the moving bed included work on milk neutralization using a moving anion bed loop. At the beginning of this project, acid removal from milk by use of a weak base resin was considered as a possibility for milk neutralization. Because of their low degree of ionization in the free base form, the weak base anion resins show little or no salt-splitting capacity. Therefore, since a weak base anion resin could remove HCl from milk with much greater efficiency than it removes citric acid from milk, HCl was used in the milk acidification process for some of the strontium removal studies. This would be significant, however, only if an adequate procedure for neutralization with a moving-bed loop could be developed. Thus far, in preliminary laboratory studies, problems of salt balance were noted in the final milk product when loop neutralization was attempted. If this procedure could be adequately developed, it would eliminate the addition of potassium hydroxide to the final product for neutralization.

6. ENGINEERING EVALUATION OF THE MOVING-BED UNIT

Many mechanical difficulties and equipment malfunctions were encountered during the initial setup procedures. Several valves leaked, resins screens were plugged and some were broken, the solubridges did not operate properly (the interface probes were insensitive), the milk pump was not adequate and the pH control was erratic.

Although the basic functions of the moving bed system have been described in section 3, some are repeated here to clarify the problem areas. The leaking valves were the Buna-N butterfly type, located between the four main compartments of the loop (fig. 2). This valve seat material is very susceptible to chafing, which results in excessive leaks. The only solution presently known is valve seat

replacement. This requires the process to be stopped, the columns emptied of resin and water, and the valve completely removed. The unit was not operated long enough to establish the life expectancy of a valve seat. It is, however, estimated to be approximately 500 to 1000 operating hours. A more abrasive-resistant resin valve seat that also meets sanitary standards is needed. Chemical Separations Corporation (CSC) tested neoprene and hypalon type seats and had little success with either.

The resin screens which permit liquid to flow through are designed to retain the resin in the unit and are located in 10 sites in each loop. All screens in the anion loop were plugged with resin and six were broken. This was apparently due to the fines of the IR 93 resin used in the test runs at the CSC. Four of the screens in the cation loop were broken and had to be rewelded. Apparently the screen-clogging problem is not a factor in the cation loop when Dowex 50W-X8 resin is used. Once the screens were cleaned and rewelded no further problems were encountered.

Conductivity probes activate three solubridges which are wired to control the opening and closing of valves in a set sequence. If for some reason these probes become insensitive, the solubridges do not function and the process cannot be controlled. Two reasons for probe insensitivity are milk fat and air pocket buildup around the probes. Solutions to these problems would be the use of different conductivity probes provided with screens located in a vertical rather than horizontal section of the loop. Possibly a more sophisticated milk-water interface control system is needed.

Air and water pockets formed around the probes because the resin bed is fluffed up and the resin beads are loosely suspended during each pulse causing a resin void. This was partially controlled with a manually operated bleeder valve. This resin displacement can be eliminated by unfolding the loop.

During the first test run with milk, an extreme pressure drop was experienced through the cation and anion milk sections. Subsequent testing revealed that an undersized relief valve on the milk pump was a contributing factor to this problem. It was also established that one milk pump was not capable of pumping milk at the designed rate (1.66 gpm) through the entire system. Therefore, a surge tank was installed between the fixed bed and the loops. Another milk pump was added to pump milk from the surge tank through the loops. This revision apparently solved the pressure drop problem.

When testing was started it became apparent something was wrong with the pH control units. It was discovered that the cation and anion reference probe contained some foreign matter and the anion reference probe was also defective. These probes were cleaned and the anion probe was replaced. Subsequent testing revealed wide pH fluctuations of the product. This pH instability was soon traced to the acid injection system. In the initial design, the acid flow continued during the pulse mode even though the milk flow had stopped, and this lowered the pH excessively. This condition was corrected by installing an electrically controlled three-way acid valve designed to divert the acid flow back to the holding tank during a pulse. A line mixer was also added at the acid injection point.

Some less serious problems and their solutions were:

- Two air solenoid valves controlling two resin valves were not sealing properly and had to be dismantled and cleaned.

- The cation strip pump was found to be [illegible]ing. The PVC discharge housing was cracked and had [illegible]e replaced.

- In the cation loop one of the delay timers was improperly wired; it contained defective wiring. This caused the strip rinse input to be inoperable.

- Both pH meter recorder-controllers, cation and anion, needed internal realignment.

- Cation resin supplied by Chemical Separations Corporation appeared to be a mixture of Dowex 2X-8 and Dowex 50W-X8 anion and cation. Subsequent laboratory tests verified that there was more Dowex 2X-8 than Dowex 50 (it was supposed to be Dowex 50). These resins were replaced.

- Both anion and cation panels had to be rewired so that the regenerant pumps and the milk pump would shut off during the pulse cycle. This required a tremendous amount of effort on our part since we were not familiar with the wiring system; and, the wiring diagrams furnished by Chemical Separations Corporation were not up to date.

- In the anion loop the milk slip temperature probe was not operating properly and a new probe was installed.

To discuss some of these problems, a Project Review Meeting on the Removal of Radionuclides from Milk and Status of the Moving Bed Ion Exchange System was held at the Southeastern Radiological Health Laboratory, Montgomery, Alabama, on October 10 and 11, 1968. The meeting was also for the purpose of exchanging information on milk decontamination technology and to discuss status of the moving bed unit, as well as to solicit comments on specific modifications to the moving bed process.

An abstract of this meeting is attached in Appendix A.

In summary, we encountered many problems in the pilot plant evaluation. Considerable modifications were required. We have not, however, altered the basic design of the pilot plant; i.e., a continuous counter-current system using water pressure to pump the resin. However, "continuous" is not literally descriptive of the system, since the input-output functions are stopped during each pulse.

7. RECOMMENDATIONS

Some changes suggested in the basic design and the reasons are as follows:

(1) Change the configuration of the moving bed loop. There are two possibilities for consideration: (a) Unfolding the loop would definitely eliminate the resin displacement problem. The column height would be increased but this would not affect the portability as the upper section could be removed during transportation. (b) Folding the loop symmetrically would theoretically correct the resin displacement problem and would also keep the column height to a minimum.

(2) Provide resin flush-out ports conveniently located on both loops. Ports are provided on the present unit, but at inaccessible locations. With the present design it is impossible to pulse all the resin out of the lower sections during clean-up.

(3) Install a resin valve between the regenerant-rinse and regenerant-strip sections. This would eliminate the solution channeling problem that was encountered during the runs, reduce regenerant dilution and increase the stripping efficiency.

(4) Place flow indicators on the milk stream. The flowrate is presently determined manually.

(5) Since neutralization has not been successfully applied to date, provisions should be made for a KOH injection system similar to the one used during the fixed bed studies at Producers' Creamery. In the event loop neutralization becomes practical, the milk working section and the NaOH input section should be placed closer together. In the present system, these sections are so far apart that a change in the NaOH flowrate does not affect the milk pH until approximately 2 hours after a change is made.

Areas for research needing further investigation with the moving bed unit are:

(1) Additional studies on cesium removal. It is not possible to maintain a high cesium removal using the present fixed bed system. Cesium removal, however, should be much more efficient with the moving bed (since fresh resin is constantly supplied) than with the fixed bed systems where the breakthrough point occurs much earlier for cesium than for strontium.

(2) Milk neutralization with a moving bed loop. This could eliminate the necessity of adding potassium hydroxide to the final product for neutralization, thus eliminating significant quantities of radioactive potassium being added to the milk as is the case with the fixed bed system.

(3) Radioisotope removal without milk acidification. This may be accomplished by the use of two moving bed loops — one in the sodium form and the other in the mixed salt form.

(4) To establish thermal and chemical conditions necessary to insure a minimum bacterial buildup.

(5) To evaluate the mobility of the unit.

(6) Nutritional and chemical evaluation of the end product. It is imperative that sufficient feeding studies and adequate chemical analysis be performed on the finished product necessary to acquire FDA approval of the system.

(7) Application to commercial milk plant operation. Much information has been accrued concerning the capabilities of the moving bed ion exchange system for the decontamination of milk; however, the question as to whether or not this system would be applicable to a commercial operation should be answered.

(8) Comparison with the fixed bed system. Because of the basic design and configuration of the moving bed loops, and since testing has been terminated, it is impossible at this time to compare this system with the fixed bed unit. If such a comparison is to be made, additional research would be necessary, and the moving bed unit should be taken to the same degree of completion as that of the fixed bed.

Some of the information presented in this document was taken from previously published articles. This has been necessary in order to show the Bureau's total activities in milk decontamination by the various processes.

For those who may be interested in additional aspects in a specific area of milk decontamination, some pertinent articles in addition to those already referenced are listed in the bibliography.

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APPENDIX A

SUMMARY OF SEMINAR PROCEEDINGS

A technical review meeting was held October 10-11, 1968, at the Southeastern Radiological Health Laboratory, Montgomery, Alabama, for the purpose of reviewing the moving bed ion exchange system for decontamination of milk.

Attendees:

Mr. Irwin R. Higgins
Dr. Gopal Murthy
Mr. Roger W. Dickerson, Jr.
Mr. William Stroup
Dr. Locke Edmondson
Mr. J. G. Terrill, Jr.
Mr. Charles L. Weaver
Mr. Ronald E. Bales
Mr. David G. Easterly
Mr. James K. Hasuike
Mrs. Jeanie Brooks
Mr. Charles R. Porter
Dr. John E. Regnier, presiding

The following is a brief summary of the recommendations and conclusions reached by the participants with regard to the future status of the project.

1. Continue evaluation of the Higgins Unit through this fiscal year. This should include:

   (a) Spiked milk studies to establish the specific conditions necessary to obtain maximum removal of iodine, strontium, cesium, and barium, and yet retain a normal balance of the salts and pH.

   (b) Establish conditions necessary for the control of microbiological contaminants.

2. Make neutralization studies with a laboratory scale unit.

3. Prepare an instruction manual for the operation of this unit.

4. Store the unit on a trailer for future use.

It was noted that in order to complete items 2, 3, and 4, it would be necessary to continue this work into FY-70.

As was discussed at this meeting, FDA approval has not as yet been obtained for this ion exchange process. The effort expended thus far would be of little value, or at least greatly minimized, unless FDA approval is obtained. This approval may require additional nutrition studies.

With this in mind, it was proposed to install the Lebanon fixed-bed unit at the SERHL for use in additional nutrition studies. The approach should be:

1. Consult with FDA in an attempt to determine what tests must be conducted in order to obtain FDA approval for the use of this process on an emergency basis.

2. These studies should include feeding studies as well as analysis of the finished product to determine any gross change in the processed milk. It is hoped that this phase of the research can be conducted jointly with Auburn University by contract.

The fixed-bed ion exchange unit should be retained at the Southeastern Radiological Health Laboratory as a model for use in the event of a radiological emergency situation.

A considerable amount of discussion was devoted to technical difficulties encountered in operation of the moving bed unit and their potential solution. These problems and solutions are listed below:

PROBLEMS	SOLUTIONS
1. Resin displacement	Open milk rinse inlet valve to drain two seconds prior to the end of pulsing or unfold the loop, use dual pulse and flow control.
2. Valve wear.	No simple solution. Change valves to ball or standard squeeze crane type valve.
3. Plugging and cleaning resin screens.	Use conical holes in screens and provide flush out.
4. Bacteriological buildup anion.	Use another type of resin which will withstand sterilization.

	PROBLEMS	SOLUTIONS
5.	Milk interface control.	Change to a properly designed screen probe.
6.	Occasional dry plug of resin between the milk rinse and regeneration sections.	Unfold loop.
7.	Service line valves shutoff.	Use ball valve solution diaphragm valve or solenoid valves power assistance for both opening and closing.

The ABSTRACT CARDS below are designed to facilitate document retrieval using COORDINATE INDEXING. They provide space for an accessing number (to be filled in by the user), suggested keywords, bibliographic information, and an abstract.

The Coordinate Indexing concept of reference material filing is readily adaptable to a variety of filing systems. Coordinate Indexing is described in the publication "IBM Data Processing Techniques - Index Organization for Information Retrieval" (C 20-8062). Copies are available through IBM Branch Offices.

The cards are furnished in triplicate to allow for flexibility in their use (for example, author card index, accession number card index).

D. G. Easterly, I. B. Brooks, J. K. Hasuike, and C. L. Weaver: DEVELOPMENT of ION EXCHANGE PROCESSES for the REMOVAL of RADIONUCLIDES from MILK. Environmental Protection Agency, Radiation Office Publication No. RO/EERL 71-1 (January 1971) 33 pp. (limited distribution).

Accession No.

ABSTRACT: Work has been conducted on the removal of radionuclides from milk by ion exchange from the late fifties to June 1969. This has progressed from laboratory-scale to a full-scale commercial process where the system was demonstrated to be feasible using fixed-bed ion exchange columns.

For three full-scale combined anion-cation runs, an average of 99.3 percent of iodine-131 and 94.6 percent

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of strontium-85 was removed. This process resulted in an increase in the total solids, ash, potassium, and citrate ions. The taste of the treated milk compared favorably with untreated milk, although some decrease in flavor score was noted.

Good radionuclide removal was obtained using the pilot plant moving-bed system. These studies, although not extensive, showed 89, 99, and 99.5 percent removal for strontium, iodine, and cesium, respectively. The limited investigations of cesium removal showed this system to have much greater potential than the fixed-bed system. For complete evaluation of this system, additional research would be needed.

KEYWORDS: Cesium-137; Iodine-131; Ion Exchange; Milk Decontamination; Radionuclide Removal; Strontium-85.

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